PATENT APPLICATION
Mo-7309-US
06160-1P67

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

APPLICATION OF	)
	) GROUP NO.: 1742
PETER R. JEPSON ET AL	)
	)
SERIAL NUMBER: 10/079,286	) EXAMINER: LOIS L. ZHENG
	)
FILED: FEBRUARY 20, 2002	)
	)
TITLE: REFRACTORY METAL PLATES	)
WITH UNIFORM TEXTURE	)
	)

DECLARATION UNDER 37 C.F.R. § 1.132

I, Peter R. Jepson, residing at 21 Marsh Avenue, Newbury, MA, 01951, United States, declare as follows.

1.   I have the following technical education and experience:

a)   I am a metallurgist having studied at Cambridge University, England, from 1969 to 1972.

b)   I received the degrees of: bachelor of arts at Cambridge University in the year of 1972; and master of arts at Cambridge University in the year of 1976.

c)   I am presently employed by H.C. Starck Inc., 45 Industrial Place, Newton, Massachusetts, 02461-1951, United States, and have been so employed since October 1998, in the research department, in particular with regard to the development of novel metal-working processes.

2.   The following tests were carried out under my immediate supervision and control:

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I. Manufacture of tantalum plates

A. Comparative example

As described in the declaration of November 16, 2005 ("the November 2005 declaration"), a tantalum plate was prepared in accordance with the disclosure of *Michaluk '475* paragraph [0040] and is referred to herein as Plate 475. This plate was prepared as follows:

1.   An ingot was made by electron-beam melting. The ingot was 200 mm diameter. Note: although less than the 9 ½" preferred in '475, this diameter was sufficient to allow the subsequent processing steps.

2.   The ingot was cut in half, for the sake of convenience.

3.   The half-ingot was mechanically cleaned (machined).

4.   The half-ingot was forged flat, starting at room temperature, to make a slab, 100 mm thick.

5.   The slab was annealed in vacuum at 1370°C, for 1.5 hours.

6.   The slab was cut in half, for the sake of convenience, to make two rolling slabs.

7.   The rolling slab was rolled to a plate 7 mm thick, starting at room temperature. Rolling passes in directions both parallel to and perpendicular to the ingot axis were included.

8.   The plate was flattened, by level-rolling.

9.   The plate was annealed in vacuum at 1120°C, for 1.5 hours. Full recrystallization was achieved, and an average grain size of 30 microns.

B. Inventive example

As also described in the November 2005 declaration, a tantalum plate in accordance with Applicants' present claims, referred to herein as Plate 286, was prepared as follows:

1.   An ingot was made by electron-beam melting. The ingot was 200 mm diameter.

2.   The ingot was cut into several parts, each 500 mm long, to achieve the required length:diameter ratio.

3.   The quarter-ingot was mechanically cleaned (machined).

4.   The quarter-ingot was upset-forged to reduce its length by 35% (step 14).

5.   The forged workpiece was annealed in vacuum at 1370°C, for 1.5 hours.

6.   The annealed workpiece was forged back to 95% of its original diameter, using swaging dies, and turning as described for step 22.

7.   The workpiece was upset-forged and forged back again, in the same manner as before. In other words, steps 14 and 22 were repeated once, as described in the last sentence of para 26.

8.   The workpiece was annealed in vacuum at 1065°C, for 1.5 hours.

9.   The work-piece was forged flat, starting at room temperature, to make a rolling slab, 100 mm thick. Note: 'side-forging', as used in '286, is the same as 'flat-forging' as used in '475.

10.   The rolling slab was rolled to a plate 6 mm thick, starting at room temperature. Rolling passes in directions both parallel to and perpendicular to the ingot axis were included. Note: 'cross-rolling' is the term used in '286 for this procedure.

11.   The plate was flattened, by level-rolling. Note: this step is not mentioned in '286, because it is not essential, and does not affect the microstructure.

12.   The plate was annealed in vacuum at 1065°C, for 1.5 hours. Full recrystallization was achieved, and an average grain size of 40 microns.

II. Texture Measurements

Texture measurements were made as described in the November 2005 declaration. The discussion of measurement is repeated here, in connection with the submission of color grain-maps . These grain-maps were previously inadvertently submitted in black and white. Black and white grain maps do not permit adequate visual interpretation of the results.

Three samples were cut along the length of each plate, and the texture examined on a cross-sectional (through-thickness) face, using an EBSD method, as described in 'Microtexture Determination' by V. Randle, Maney Publishing, 2003. For each sample, a grain map was generated. The color scheme of the grain maps

is shown in Inverse Pole Figure ("IPF") format, in which the colors represent the orientation of each analyzed point, and thus each grain, relative to the plate Normal Direction ("ND"). Note, however, that what is black on the IPF is grey on the maps. The map extends from one surface of the plate (top of the map) to the other surface of the plate (bottom of the map). The thickness of Plate 475 was about 7 mm, and that of Plate 286 about 6 mm. The length of each plate sample examined, in each case (i.e., for Plate 475 and Plate 286), was about 1.5 mm.

Plate 475 and Plate 286 were in each case observed to possess some thickness variation across each plate, and thickness tapering towards the edge of each plate. In the following presentation of texture results, Samples A through C of Plate 475 show less thickness tapering than those of Plate 268, due to the variation in thickness across the plates (i.e., the three samples cut from Plate 475 happened to have a very similar thickness).

Texture Results

IPF
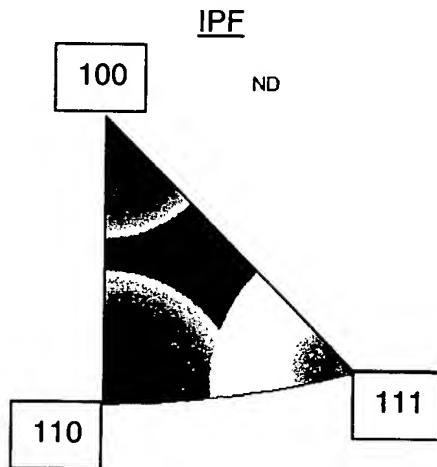
Texture Results (cont'd):

Plate 475
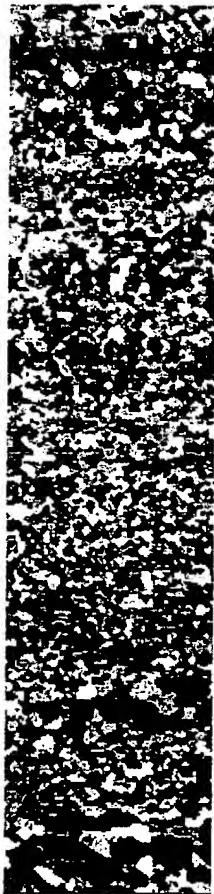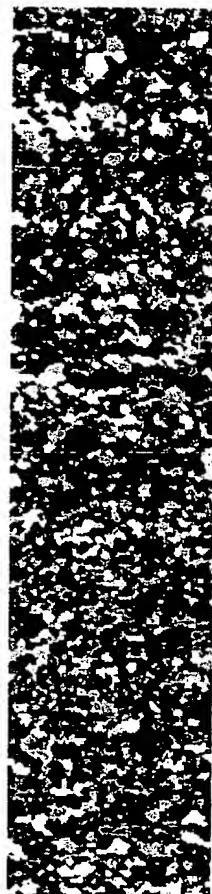
Sample A   Sample B   Sample C

Texture Results (cont'd):

Plate 286

Sample A
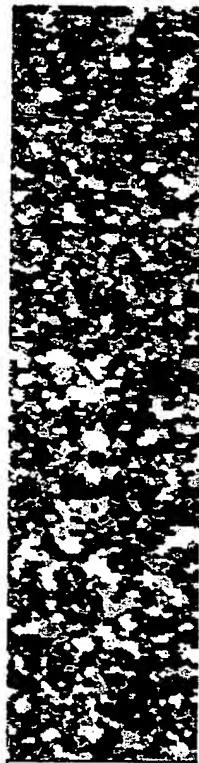
Sample B

Sample C

3.   The samples from Plate 475, when analyzed in accordance with the method described in paragraph [0030] of *Michaluk '475*, were found to have an Ln ratio of {111}:{100} center peak intensities within the same increment of greater than about -4.0, and {100} peak center intensities less than 15 random. See Table 1 for an analysis of Sample A. In fact, the lowest Ln ratio of {111}:{100} peak intensities was −1.74, and the highest {100} peak intensity was 12.5 random. Thus the teachings of Michaluk in '475 have been reproduced.

4.   Visual examination of the grain maps for Plate 475 (samples A, B and C) indicates clearly that although the measurement criteria of Michaluk '475 have been met, the texture in any one map (ie. throgh the thickness) is far from uniform or homogeneous. Horizontal bands of both yellow (111) and blue (100) can be seen. For example, there is a wide band of yellow about ¼ of the way down the map (which corresponds to the high (111) peak intensities of increments 4, 5 and 6 of Table 1).

It should also be noted that a natural log ratio of (111): (100) peak intensities of −4.0 corresponds to a ratio of peak intensities of 1:55. A texture with (111) to (100) intensity ratio of 1:55 would be described by one skilled in the art as a very strong 100 texture. Variation in texture from one point in a sample to another of 1:55 to 1:1 (equal intensities of 100 and 111 peaks) would be described by one skilled in the art as a very high level of variability, indicating that the plates are in fact substantially non-uniform. The fact that Michaluk uses the words "uniform" and "homogeneous" to describe the texture of the niobium plates provided in that reference is irrelevant in view of the measurements described therein.

5.   Various methods of measuring the texture of a metal sample are known and used in the art. Two such methods are that described in '475 (the peak intensity method), and that described in '286 (the % distribution method). Both methods were used to measure the texture of Plates 475 and 286. The details of the methods used are as follows:

'475 Method: The thickness was divided into 20 increments. For each increment, the peak intensity was calculated using a 10° half-width (which is not specified in '475, but is standard in the industry).

'286 Method: The thickness was divided into 8 increments. The number of increments to be used is not specified in '286, but 8 is typical, and the number of increments is not critical. For each increment, the % of the area within 15° of 100, and within 15° of 111, is calculated, and the difference (the distribution) is further calculated.

The results of these methods are provided in Tables 1 to 4 below, and summarized as follows:

'475 Method

	(100) peak max	In (111): (100) min
Plate 475	12.5	-1.74
Plate 286	13.7	-1.74

'286 Method

	(111-15) – (100-15) max %	(111-15) – (100-15) min %	Difference (%)
Plate 475	45	-17	62
Plate 286	-6	-31	25

It can be seen that the results of the '475 method are very similar for the two plates, and do not reflect the difference in uniformity between the plates visible in the grain maps. However the '286 method shows a much lower distribution for Plate 286 (25% as compared to 62%), and this reflects the difference between the plates visible in the grain maps, and is therefore a better method to use to quantify texture uniformity.

6.   As also shown in the measurement data, the results of the '286 method of measurement correspond directly to the language now present in Claim 1, namely, that the distribution of {100} and {111} crystallographic orientations varies by less than 30% across any thickness of the metal plate. The inventive sample meets this claim limitation, while the comparative sample of Michaluk does not.

7.   Further visual examination of the grain maps for Plate 475 indicates that the texture in Sample A is very different from that of Sample B, and that both

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are different from that of Sample C. This is not surprising, as there is no teaching in '475 concerning uniformity of texture across the plate. Visual examination of the grain maps (Samples A, B and C) for Plate 286 indicates substantial similarity between them, as would be expected in light of our claim that "said refractory metal plate has (i) a distribution of {100} and {111} crystallographic orientations that varies by less than 30% across the surface of any plane of said refractory metal plate, said planes being orthogonal to the thickness of said refractory metal plate". This can be illustrated for Plate 286, taking Increment 1 as an example:

TABLE 5: PLATE 286, SAMPLES A, B AND C, METHOD 286

SAMPLE #	100-15	111-15	DIFFERENCE
A	37	18	-19
B	48	14	-36
C	44	17	-27

Thus, the distribution (difference between max and min) is 17%, within the '< 30%' range recited in claim 1. Given that there is no mention in the Detailed Description of '475 of uniformity across a plate, and given that Plate 475 does not show uniformity across itself, the development of a plate which does show uniformity across itself (within the limits given), is clearly inventive and nonobvious in view of Michaluk.

8.   It is asserted in the Office Action that since Michaluk teaches the presence of {100} and {111} crystallographic orientations and a "uniform texture" that the texture distribution variation of the sputtering plate of Michaluk must be very small, thus falling within the claimed "less than 30% variation". This is incorrect. It is not the mere presence of {100} and {111} crystallographic orientations that provides the uniform texture; as explained above, it is the variability of the relationship between the amounts of each orientation (or, rather, a lack of variability) that indicates uniformity of texture. As explained above, Michaluk by his own account allows a very high variability. The measurements made above clearly show that a plate made by the method of Michaluk does not fall within the claimed range.

9.   Annealing temperatures for tantalum are well-known. Discussion of

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annealing temperatures can be found, for example, in "Effect of Processing Variables on Texture and Texture Gradients in Tantalum", by J.B. Clark et al, Metallurgical Transactions A, Volume 22A, Sept. 1991, p. 2039 – 2048. Typically a range of temperatures such as 900°C to 1400°C are reported as suitable, with the preferred temperature range depending on whether it is the first anneal, an intermediate anneal or the final anneal. One skilled in the art of tantalum metallurgy can easily determine the appropriate time and temperature for annealing tantalum, without undue experimentation. The uniformity of texture provided by the method of making tantalum plates in the present invention would be expected to occur across the entire range of standard times and temperatures for the various process steps, as would be understood by one skilled in the art.

9. The sputtering targets of the present invention are made by a method that is very different from the method of making targets described in Michaluk '475. This novel method provides a substantial improvement of uniformity of texture in the plate, as compared to prior art methods, including that of Michaluk. It is my well-considered opinion that the tantalum sputtering targets of the present invention are not obvious in view of the teachings of Michaluk '475.

10. I further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States code and that such willful false statements may jeopardize the validity of pending Application Serial Number 10/079,286 or any patent issuing thereon.

Signed at Newton, MA this 26th day of May, 2006.
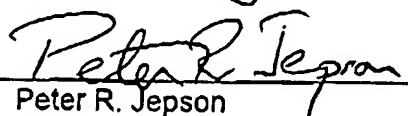
Peter R. Jepson

TABLE 1: PLATE 475, SAMPLE A, METHOD 475

INCREMENT #	100 INTENSITY	111 INTENSITY	LN(111): (100)
1	8.2	14.1	0.54
2	9.5	13.2	0.33
3	12.3	7.4	-0.51
4	5.7	15.1	0.97
5	1.5	21.4	2.66
6	1.9	12.7	1.90
7	5.1	3.1	-0.50
8	4.5	1.1	-1.41
9	4.7	2.7	-0.55
10	0.8	4.1	1.63
11	4.3	5.5	0.25
12	4.4	3.8	-0.15
13	3.3	4.5	0.31
14	5.8	3.6	-0.48
15	7.1	2.1	-1.22
16	8.6	2.5	-1.24
17	12.5	2.2	-1.74
18	5.8	2.3	-0.92
19	6.5	10.4	0.47
20	4.2	2.0	-0.74

TABLE 2: PLATE 286, SAMPLE A, METHOD 475

INCREMENT #	100 INTENSITY	111 INTENSITY	LN(111): (100)
1	7.0	3.3	-0.75
2	7.9	1.7	-1.53
3	8.8	1.9	-1.43
4	10.4	2.4	-1.47
5	9.1	2.9	-1.14
6	6.5	2.2	-1.08
7	3.7	3.9	0.05
8	5.3	4.1	-0.26
9	5.7	6.7	0.16
10	8.5	1.9	-1.50
11	6.2	2.1	-1.08
12	6.9	3.1	-0.80
13	6.4	2.2	-1.07
14	6.2	1.4	-1.49
15	5.9	4.2	-0.34
16	4.1	5.2	0.24
17	8.7	4.6	-0.64
18	13.7	2.4	-1.74
19	9.1	4.0	-0.82
20	8.9	3.1	-1.05

TABLE 3: PLATE 475, SAMPLE A, METHOD 286

INCREMENT #	100-15	111-15	DIFFERENCE
1	32	55	23
2	19	64	45
3	16	38	22
4	19	22	3
5	18	34	16
6	35	21	-14
7	31	14	-17
8	24	10	-14

TABLE 4: PLATE 286, SAMPLE A, METHOD 286

INCREMENT #	100-15	111-15	DIFFERENCE
1	37	18	-19
2	49	18	-31
3	32	26	-6
4	35	28	-7
5	40	21	-19
6	37	19	-18
7	40	30	-10
8	42	21	-21